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Scope of Research

Kinetic and mechanistic analyses are made for better understandings of the chemical and physicochemical reactions occurring in polymerization systems and for better routes to the synthesis of well-defined polymers. By various polymerization techniques, in particular, living polymerizations, new well-defined polymers or polymer assemblies are prepared, and their structure/properties relationships are precisely analyzed. Projects in progress include: (1) kinetics and mechanisms of living radical polymerization (LRP). (2) Synthesis of new polymeric materials by living polymerizations and their structure/properties studies. (3) Synthesis, properties, and applications of high-density polymer brushes (HDPB).

Research Activities (Year 2003)

Presentations

2 presentations, The 1st Trilateral Workshop on Organic Chemistry, Taiwan, 17 - 19 Jan.

Structure and Properties of HDPB (Invited Lecture), Fukuda T, and 2 other presentations, The 2nd NIST-KIPS Symposium on Polymer Science, USA, 20 - 21 Mar.

Structure and Properties of HDPB (Invited Lecture), Tsujii Y, and 3 other presentations, 225th ACS National Meeting, USA, 23 - 27 Mar.

Formation of Branched Polymer Systems by LRP (Invited Lecture), Fukuda T, and 1 other presentation, 9th International Seminar on Elastomers, Kyoto, 2 - 4 Apr.

Synthesis and Properties of Newly Developed Polymer Brushes (SPSJ Willey Award Lecture), Tsujii Y, Soc. Polym. Sci., Jpn., 25 Sep.

8 presentations, Spring Meeting, Soc. Polym. Sci., Jpn., 28 - 30 May.

3 presentations, Polymer Symposium, Soc. Polym. Sci., Jpn., 10 Jul.

8 presentations, Autumn Meeting, Soc. Polym. Sci., Jpn., 24 - 26 Sep.

3 presentations, Annual Meeting, Soc. Fib. Sci. Tech., Jpn., 11 - 13 Jun.

Grants

Fukuda T, Science and Technology of HDPB, Grant-in-Aid for Science Research (A)(2), 1 Apr 2002 - 31 Mar 2005.

Tsujii Y, Fundamental Study on HDPB, Grant-in-Aid for Science Research (B)(2), 1 Apr 2002 - 31 Mar 2005.

Tsujii Y, Nanostructure Control by Mixed Polymer Brushes, Grant-in-Aid for Exploratory Research, 1 Apr 2003 - 31 Mar 2005.

Ohno K, Gold Nanoparticles Coated with HDPB, Grant-in-Aid for Young Scientists (B), 1 Apr 2002 - 31 Mar 2004.

Goto A, Chain Length Dependence of k_p , Grant-in-Aid for Young Scientists (B), 1 Apr 2003 - 31 Mar 2005.

Tanaka K, Construction of Nanoelectronic Devices, CREST, Jpn. Sci. Tech. Corp., 1 Nov 2002 - 31 Oct 2007.

Kaya K, Collaboratory on Electron Correlations, Grant-in-Aid for Cre. Sci. Res., 1 Apr 2001 - 31 Mar 2006.

Award

Tsujii Y, SPSJ Willey Award for 2003, Synthesis and Properties of Newly Developed Polymer Brushes, Soc. Polym. Sci., Jpn., 25 Sep 2003.

Fabrication of Ordered Arrays of Gold Nanoparticles Coated with High-Density Polymer Brushes

By means of surface-initiated LRP, we have synthesized a metal-polymer hybrid which has the core of the gold nanoparticle (AuNP) prepared by a simple one-pot method with a narrow size distribution, and the shell of the poly(methyl methacrylate) (PMMA) end-grafted on the core surface with a surface density as high as about 0.4 chains/nm² and a controlled chain length and chain length distribution. Primarily due to the high surface density, the PMMA grafts at the air-water interface exert interparticle interactions of an extremely long range, a range comparable to the full length of the PMMA chain. As a result, we have succeeded for the first time in fabricating a two-dimensional array of the AuNPs with a high degree of structural order and an exceptionally wide controllability of the interparticle distance (Figure 1). Owing to the simplicity and versatility of surface-initiated LRP, this work may be extended to other metal and semiconductor nanoparticles for their 2D and 3D ordered assemblies with wide controllability of lattice parameters.

K. Ohno, K. Koh, Y. Tsujii, T. Fukuda, *Angew. Chem. Int. Ed.*, **42**, 2751-2754 (2003). This paper was awarded the status of a Hot Paper by *Angewandte Chemie*.

Precision Synthesis of Organic/Inorganic Hybrid Nanocapsule with Silanol-Functionalized Micelle Template

A novel technique is presented for the construction of a silica-based capsule of tens of nanometers size (Figure 2). An amphiphilic block copolymer carrying several protected silanol groups near the free end of the hydrophilic block was synthesized by the copper-mediated LRP (ATRP) in conjunction with the sequential monomer addition technique. In an aqueous media, the amphiphile formed a micelle with a diameter about 40 nm with the silanol groups populated near the micelle surface. Mixing the micelle with an active silicate solution at pH 9 formed a silica layer with a thickness of several nanometers on the micelle surface, thus producing a new type of organic/inorganic hybrid nanocapsule composed of the core of a polymeric micelle and the shell of a silica thin layer (Figure 3).

K. Koh, K. Ohno, Y. Tsujii, T. Fukuda, *Angew. Chem. Int. Ed.*, **42**, 4194-4197 (2003). This paper was awarded the status of a Very Important Paper by *Angewandte Chemie*.

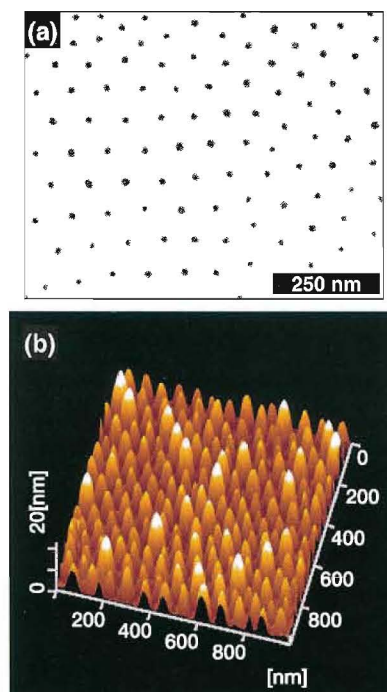


Figure 1. (a) TEM and (b) AFM micrographs of the transferred films of AuNP-PMMA hybrid: M_n of the PMMA graft = 28 000.

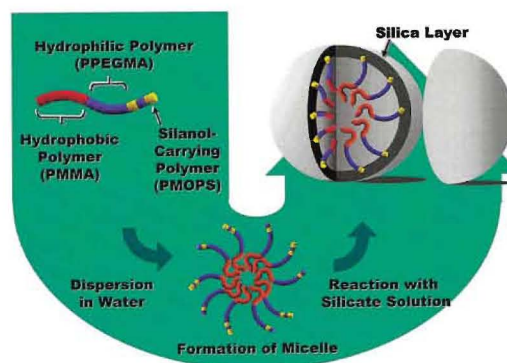


Figure 2. Strategy for the synthesis of an organic/inorganic hybrid nanocapsule.

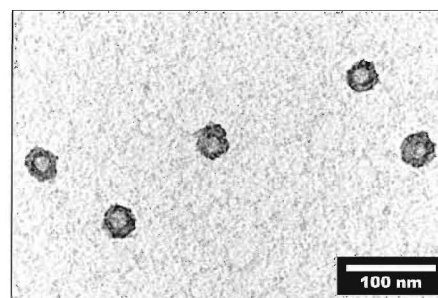


Figure 3. TEM image of the organic/inorganic hybrid nanocapsule.